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Roodbeen

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(54) SYSTEM AND METHOD FOR SEPARATING A GAS MIXTURE

(71) Applicant: **Haffmans B.V.**, Venlo (NL)

(72) Inventor: **J. E. Roodbeen**, Venlo (NL)

(73) Assignee: **HAFFMANS B.V.**, Venlo (NL)

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CPC ***B01D 53/226*** (2013.01); ***B01D 53/227*** (2013.01); *B01D 2256/245* (2013.01); *B01D 2257/504* (2013.01); *B01D 2258/05* (2013.01); *B01D 2317/025* (2013.01)

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See application file for complete search history.

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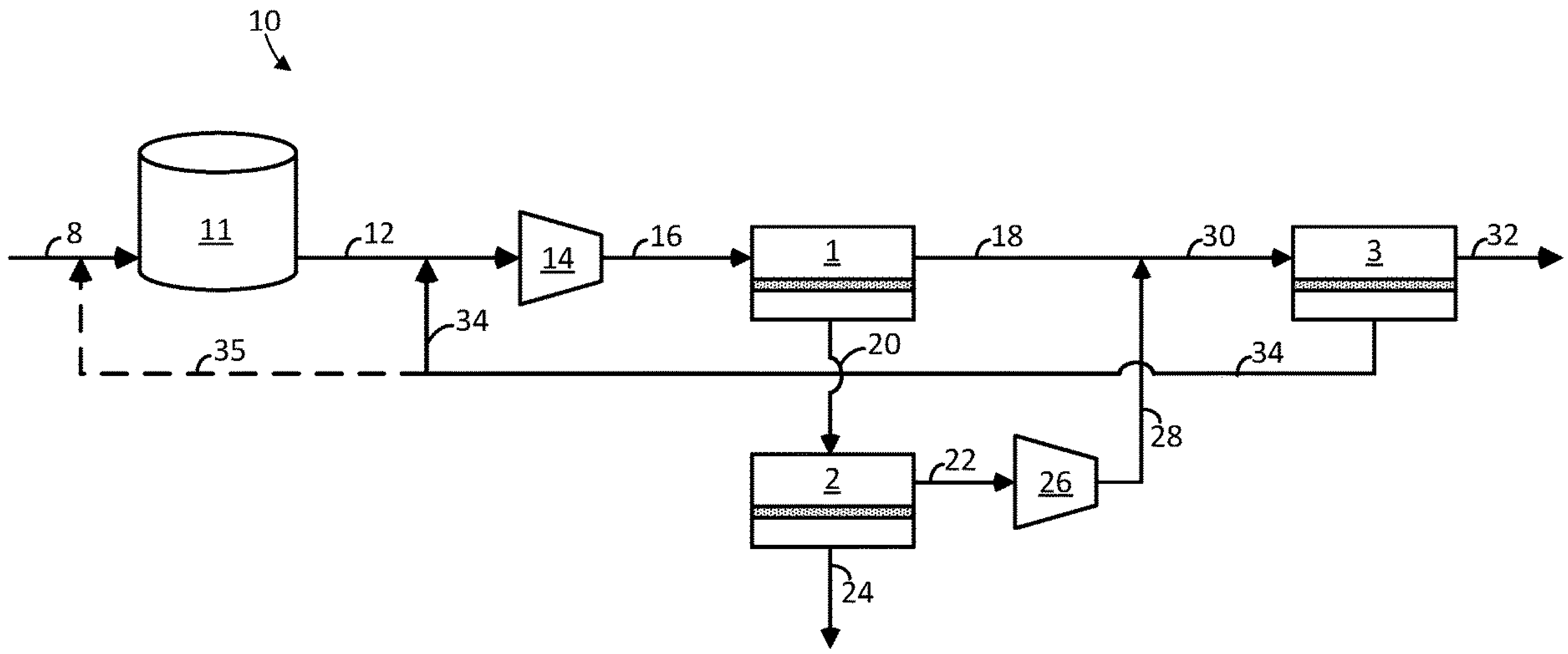
Primary Examiner — Jason M Greene
(74) *Attorney, Agent, or Firm* — Husch Blackwell LLP

(57) **ABSTRACT**

An apparatus and process for separating a gas mixture is disclosed. The apparatus includes a first membrane stage, a second membrane stage, and a third membrane stage. The first membrane stage includes a first gas separation membrane configured to separate the gas mixture into a first retentate stream and a first permeate stream. The second membrane stage includes a second gas separation membrane configured to separate the first permeate stream into a second retentate stream and a second permeate stream. The second retentate stream of the second membrane stage is recycled back to connect with the first retentate stream to form a mixed fluid stream. The third membrane stage includes a third gas separation membrane configured to separate the mixed fluid stream into a third retentate stream and a third permeate stream, and the third retentate stream is configured to be withdrawn as a product or discarded.

18 Claims, 5 Drawing Sheets



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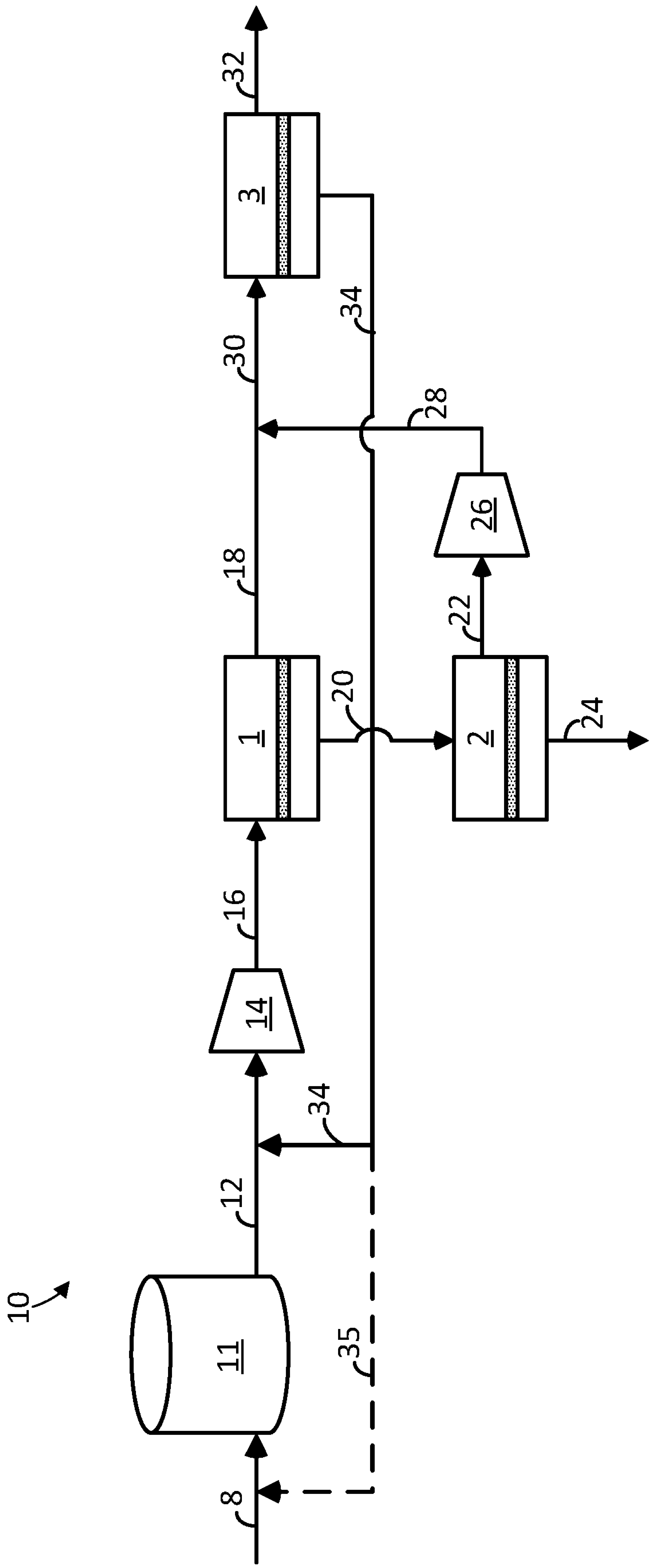

FIG. 1

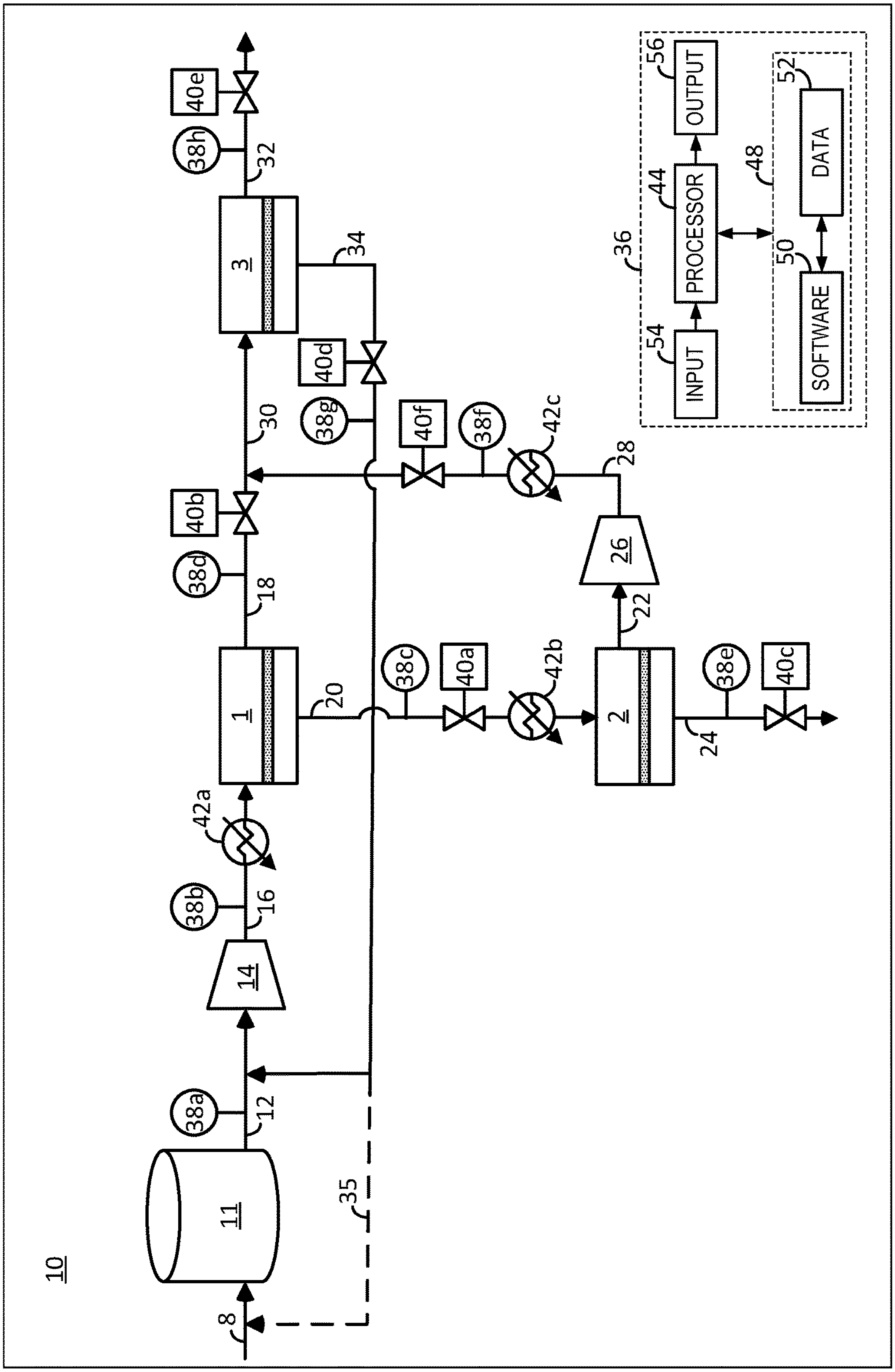
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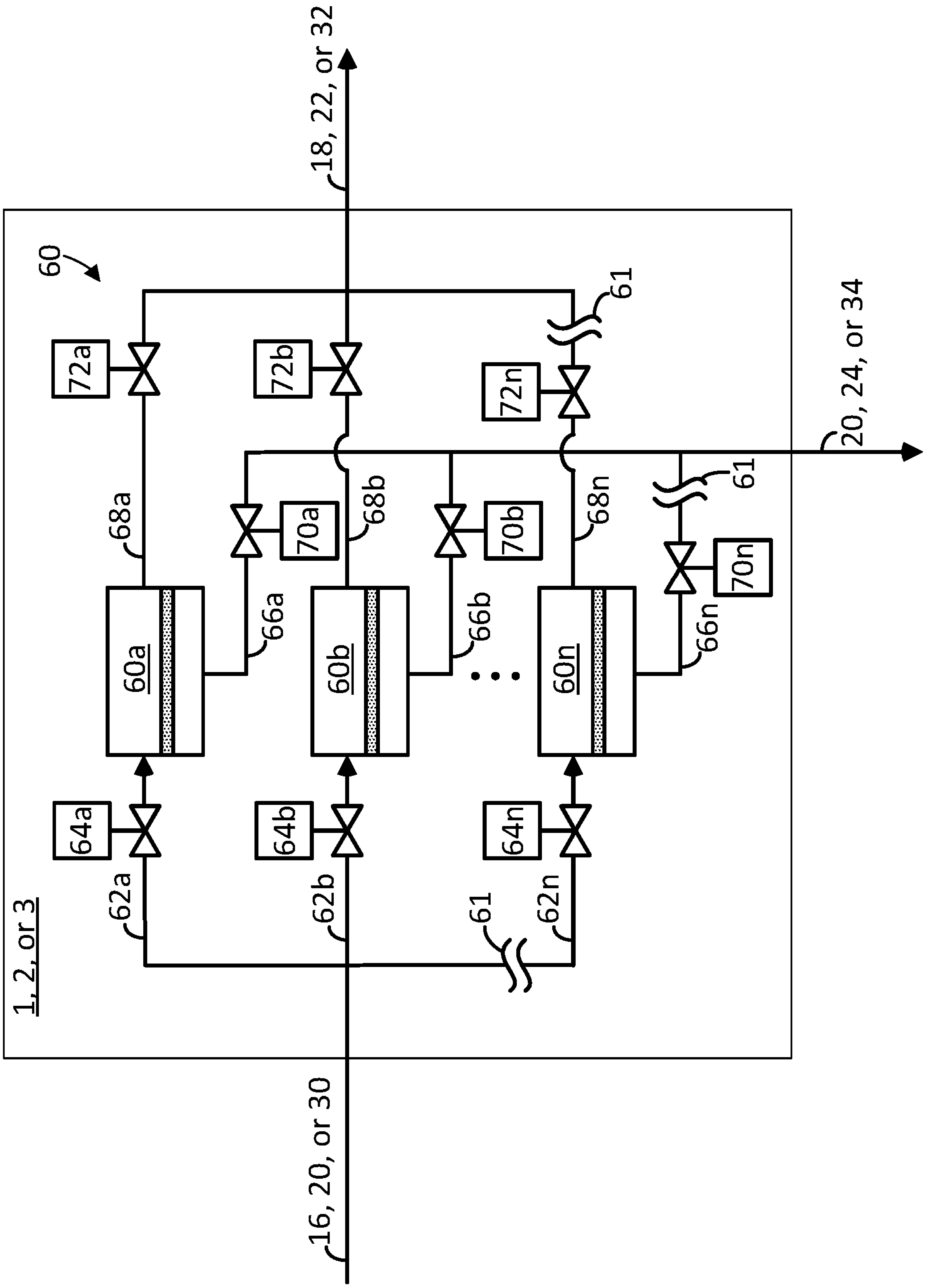
FIG. 2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16, 20, or 30

FIG. 3

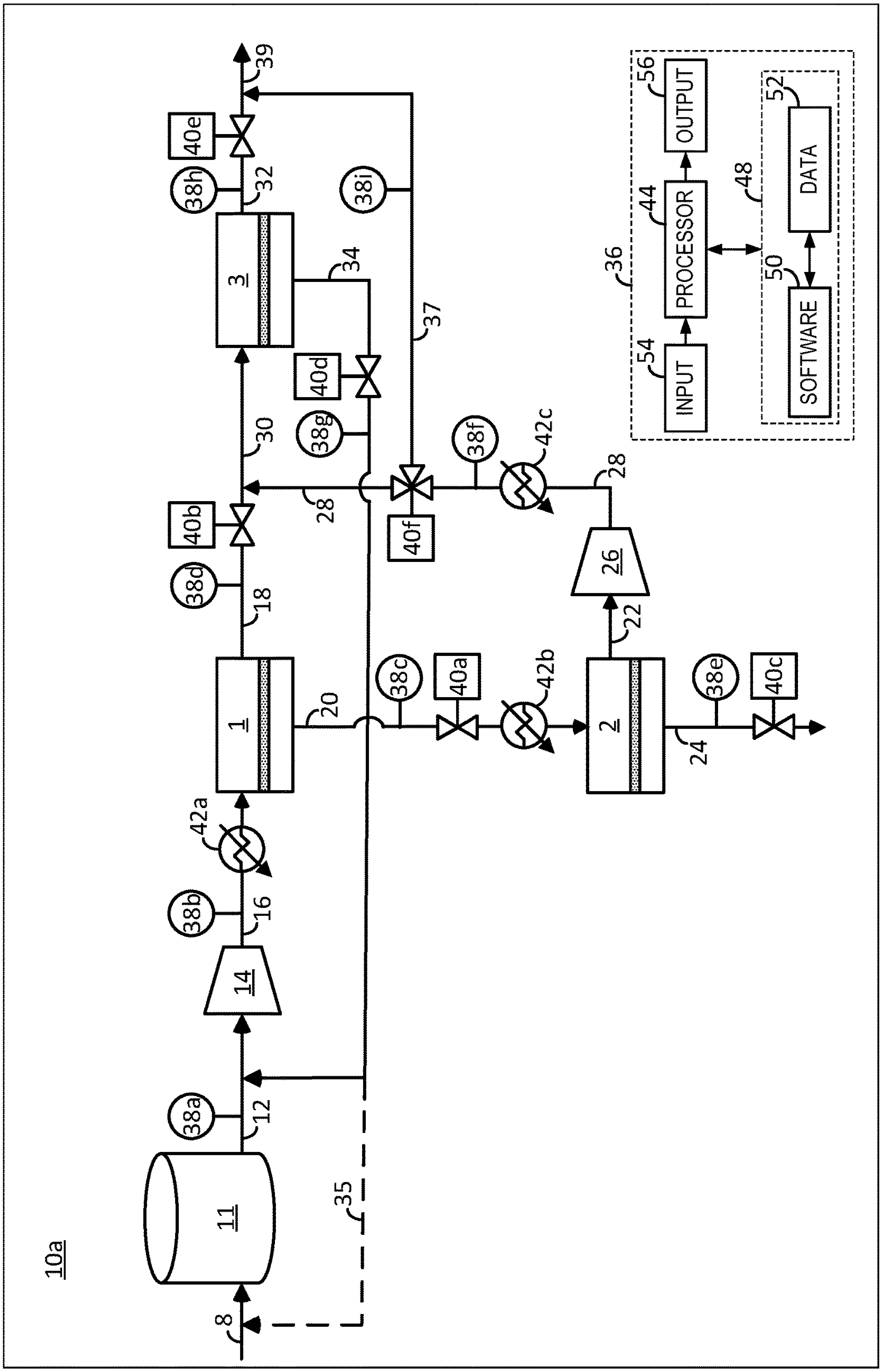
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FIG. 4

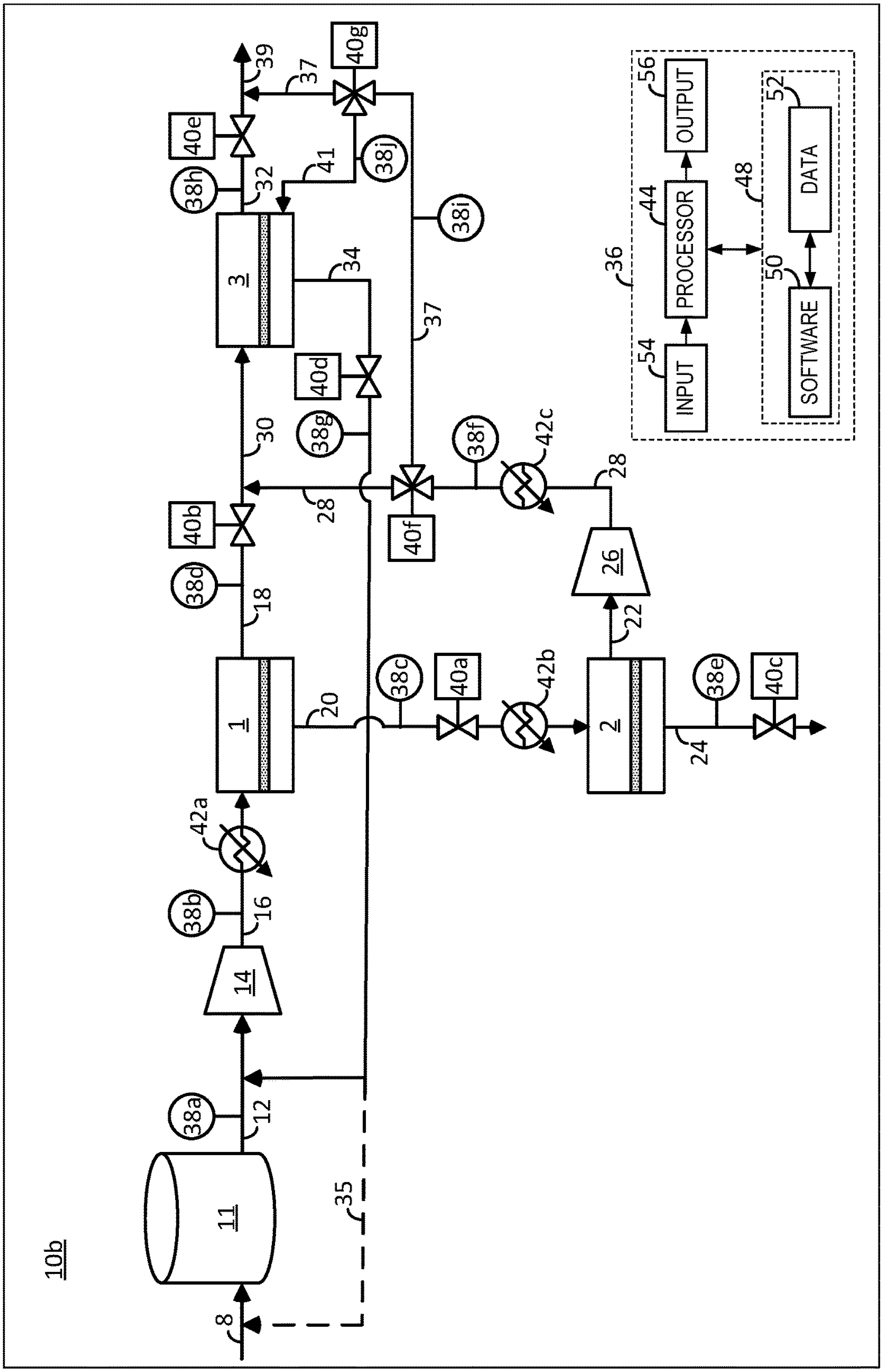
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FIG. 5

SYSTEM AND METHOD FOR SEPARATING A GAS MIXTURE

CROSS-REFERENCE TO RELATED APPLICATIONS

The instant application claims priority to U.S. Provisional Patent Application No. 62/804,338, filed on Feb. 12, 2019. The afore-mentioned patent application is incorporated by reference in its entirety herein.

BACKGROUND

Polymeric gas separation membranes are well known and commonly used in industry to purify, upgrade, and/or remove a desired component from gas mixtures. Common industrial uses for gas separation membranes include the recovery of hydrogen from gas mixtures (e.g., ammonia or oil refinery process streams), the recovery of methane from gas mixtures (e.g., natural gas or biogas), the production of oxygen-enriched air, and/or the removal of impurities from gas streams (e.g., removal of water vapor, volatile organic compounds, and H_2S).

Recently, there has been an increased interest in separation systems that separate methane from biogas or natural gas streams. Methane, especially biomethane, is a desirable product as it can be used in a variety of commercial applications, such as a source of energy for heating, for the production of chemicals, or for use as a fuel, among others. However, efficiently separating methane from carbon dioxide is a non-trivial task owing to the low selectivity of polymeric membrane materials to these gases. In order to counteract the low single-stage selectivity, commercial systems typically utilize three or more membrane stages operating at high pressures to achieve the desired purity.

Gas separation at an industrial scale is challenging. Unlike laboratory conditions that are typically performed using pure gas samples at modest temperatures and pressures, industrial conditions include exposing the membranes to multi-component gas streams having contaminated liquid and/or solid impurities, while operating at elevated temperatures and pressure. Often times, the gas source provides the gas mixture to the separation system irregularly, for example. The gas mixture typically fluctuates both in composition and flow rate over time, making it difficult to control the gas separation system to reliably and consistently produce a desired purity of the product gas stream.

Currently, there remains a need for improvements to existing commercial gas separation processes to develop more cost effective and energy efficient systems that exhibit superior process control, especially for processes directed to separating methane from biogas and natural gas streams.

SUMMARY OF THE DISCLOSURE

The present disclosure addresses the aforementioned drawbacks by providing a process for separating a gas mixture using a multi-stage membrane separation system having reduced operational costs and improved separation efficiency over conventional systems.

Some embodiments of the disclosure provide an apparatus for separating a gas mixture having two or more components. In particular, the apparatus includes a plurality of membrane separation stages including a first membrane stage, a second membrane stage, and a third membrane stage. The first membrane stage includes a first gas separation membrane configured to separate the gas mixture into a first retentate stream and a first permeate stream. The second membrane includes a second gas separation membrane configured to separate the first permeate stream into a second retentate stream and a second permeate stream. The second retentate stream of the second membrane stage is placed in fluid communication with the first retentate stream to form a mixed fluid stream. The third membrane stage includes a third gas separation membrane configured to separate the mixed fluid stream into a third retentate stream and a third permeate stream, wherein the third retentate stream is configured to be withdrawn as a product or discarded.

In some embodiments, the apparatus includes a gas source comprising the gas mixture, where the gas source is configured to be in fluid communication with the first membrane stage. The gas mixture in the gas source may comprise biogas.

In further embodiments, the apparatus includes a first gas transport device having a suction side and a discharge side. The suction side may be connected to an inlet gas line that places the gas source in fluid communication with the first gas transport device, where the discharge side is connected to a feed gas line that places the first gas transport device in fluid communication with the first membrane stage.

In some embodiments, the third permeate stream of the third membrane stage is recycled and provided to the inlet gas line.

In further embodiments, the apparatus further includes a second gas transport device having a suction side connected to the second retentate stream and a discharge side connected to a continuing stream, wherein the continuing stream places the first retentate stream in fluid communication with the second gas transport device. The first gas transport device and the second gas transport device may comprise a compressor or a blower. In some embodiments, the first gas transport device and the second gas transport device are sized such that the mass flowrate of gas transported through the first gas transport device to the second gas transport device is a ratio of 10:1 to 100:1, or of about 10:1 to about 100:1.

In some embodiments, the first gas separation membrane, the second gas separation membrane, and the third gas separation membrane have a selectivity of carbon dioxide/methane that is greater than 3, or is greater than about 3. In some embodiments, the first gas separation membrane, the second gas separation membrane, and the third gas separation membrane each have a carbon dioxide/methane selectivity that is different with respect to each other. In further embodiments, the first gas separation membrane, the second gas separation membrane, and/or the third gas separation membrane are characterized as having a selectivity of carbon dioxide/methane that ranges between 20 to 75, or between about 20 to about 75. In some embodiments, the first gas separation membrane, the second gas separation membrane, and/or the third gas separation membrane are characterized as having a selectivity of carbon dioxide/methane that ranges between 30 to 60, or between about 30 to about 60.

In further embodiments, at least one of the first membrane stage, the second membrane stage, and the third membrane stage includes two or more gas separation membranes configured in a parallel arrangement. Additionally, or alternatively, at least one of the first membrane stage, the second membrane stage, and the third membrane stage includes two or more gas separation membranes configured in a series arrangement.

In some embodiments, the apparatus further includes a feed line measuring device configured to acquire one or more process parameters associated with the inlet gas line and/or in the feed gas line. The apparatus may additionally include a first gas transport device having a suction side and a discharge side, where the suction side is connected to an inlet gas line that places the gas source in fluid communication with the first gas transport device. The discharge side may be connected to a feed gas line that places the first gas transport device in fluid communication with the first membrane stage. In some embodiments, the apparatus further includes a controller in electrical communication with the first gas transport device and the feed line measuring device. The controller may be configured to execute a stored program to: adjust a speed of rotation of the first gas transport device to obtain a desired pressure set-point in the feed gas line, where the speed of rotation of the first gas transport device is adjusted in response to a change in the one or more process parameters in the inlet gas line and/or in the feed gas line.

In further embodiments, the feed line measuring device includes a flowrate measuring device and/or a pressure measuring device and/or a composition measuring device.

In some embodiments, the apparatus further includes a first retentate measuring device configured to acquire one or more process parameters associated with the first retentate stream, and a second gas transport device having a suction side connected to the second retentate stream and an discharge side connected to a continuing stream, where the continuing stream places the first retentate stream in fluid communication with the second gas transport device. The apparatus may include a controller in electrical communication with the second gas transport device and the first retentate measuring device. The controller may be configured to execute a stored program to: adjust a speed of rotation of the second gas transport device to obtain a desired pressure set-point in the continuing stream, where the speed of rotation of the second gas transport device is adjusted in response to a change in the one or more process parameters associated with the first retentate stream.

In further embodiments, the first retentate measuring device comprises a pressure measuring device, and wherein the controller is further programmed to adjust the speed of rotation of the second gas transport device such that the pressure of the continuing stream is substantially the same as the pressure of the first retentate stream.

In some embodiments, the apparatus further comprises a third retentate measuring device configured to acquire one or more process parameters associated with the third retentate stream and/or a second permeate measuring device configured to acquire one or more process parameters associated with the second permeate stream. In some embodiments, the apparatus further includes a process control device configured to increase or decrease the pressure of the first membrane stage, the second membrane stage, or the third membrane stage. A controller may be placed in electrical communication with the third retentate measuring device, the second permeate measuring device, and the process control device. The controller may be configured to execute a stored program to: adjust the pressure of the first membrane stage, the second membrane stage, or the third membrane stage to a desired pressure set-point in response to a change in the one or more process parameters associated with the third retentate stream and/or the second permeate stream. In some embodiments, the third retentate measuring device and/or the second permeate measuring device includes a composition measuring device.

In further embodiments, a process for separating a gas mixture having two or more components is disclosed. In particular, the process includes feeding the gas mixture to a first membrane separation stage to separate the gas mixture into a first retentate stream and a first permeate stream. The first permeate stream of the first membrane stage is fed to a second membrane separation stage where the first permeate stream is separated into a second retentate stream and a second permeate stream. The second retentate stream is then recycled back to the first retentate stream of the first membrane separation stage to form a mixed fluid stream.

In some embodiments, an apparatus for separating a gas mixture having at least two components is disclosed. In particular, the apparatus comprises a plurality of membrane separation stages including a first membrane stage, a second membrane stage, and a third membrane stage. In some embodiments, the first membrane stage has a first gas separation membrane configured to separate the gas mixture into a first retentate stream and a first permeate stream, the second membrane stage has a second gas separation membrane configured to separate the first permeate stream into a second retentate stream and a second permeate stream, and the third membrane stage has a third gas separation membrane configured to separate the first retentate stream into a third retentate stream and a third permeate stream. In some embodiments, the second retentate stream is placed in fluid communication with the third retentate stream to form a mixed retentate stream that is collected as product, discarded, or further processed.

In further embodiments, the second retentate stream of the apparatus is split into a first continuing stream and a second continuing stream, where the first continuing stream is placed in fluid communication with the first retentate stream, and the second continuing stream is placed in fluid communication with the third retentate stream to form the mixed retentate stream.

In some embodiments, a valve is configured to regulate the flowrate between the first continuing stream and the second continuing stream. In further embodiments, the apparatus further includes a controller in electrical communication with the valve, the controller executing a stored program to: adjust the flowrate or pressure of the first continuing stream and the second continuing stream by moving the valve between an open and closed position.

In some embodiments, the apparatus further includes a third retentate measuring device configured to acquire one or more process parameters associated with the third retentate stream, wherein the controller is in electrical communication with the third retentate measuring device. The controller may execute a stored program to: adjust the flowrate or pressure of the first continuing stream and the second continuing stream based on the one or more process parameters in the third retentate stream.

In further embodiments, an apparatus for separating a gas mixture having at least two components is disclosed. In particular, the apparatus comprises a plurality of membrane separation stages including a first membrane stage, a second membrane stage, and a third membrane stage. In some embodiments, the first membrane stage has a first gas separation membrane configured to separate the gas mixture into a first retentate stream and a first permeate stream, the second membrane stage has a second gas separation membrane configured to separate the first permeate stream into a second retentate stream and a second permeate stream, and the third membrane stage has a third gas separation membrane configured to separate the first retentate stream into a third retentate stream and a third permeate stream. In some embodiments, the second retentate stream is placed in fluid communication with a permeate side of the third membrane stage such that the second retentate stream is fed across at least a portion of the permeate side of the third gas separation membrane. In some embodiments, the second retentate stream is configured to discharge from the third membrane stage through the third permeate stream.

In some embodiments, the second retentate stream is split into a first continuing stream and a second continuing stream, where the first continuing stream is placed in fluid communication with the first retentate stream. In some embodiments, a first fraction of the second continuing stream is placed in fluid communication with the third retentate stream to form a mixed retentate stream and a second fraction of the second continuing stream is fed across at least a portion of the permeate side of the third gas separation membrane.

In further embodiments, the apparatus includes a valve configured to regulate the flowrate between the first fraction of the second continuing stream and the second fraction of the second continuing stream. A controller may be placed in electrical communication with the valve. The controller may be configured to execute a stored program to: adjust the flowrate or pressure of the first or second fraction of the second continuing stream by moving a position of the valve between an open and closed position.

In some embodiments, the apparatus further includes a third retentate measuring device configured to acquire one or more process parameters associated with the third retentate stream. The controller may be in electrical communication with the third retentate measuring device, and may be configured to execute a stored program to: adjust the flowrate or pressure of the first or second fraction of the second continuing stream based on the one or more process parameters in the third retentate stream.

In some embodiments, an apparatus for separating a gas mixture having at least two components is disclosed. In particular, the apparatus includes an inlet feed line configured to place a gas source comprising the gas mixture in fluid communication with a membrane separation stage having a plurality of membranes disposed therein. The plurality of membranes are configured to have a total membrane surface area, where each membrane in the plurality of membranes includes a gas separation membrane that separates the gas mixture into a retentate stream and permeate stream. The apparatus further includes a pooled permeate stream formed by combining each of the permeate streams from the plurality of membranes, where the pooled permeate stream is configured to exit the membrane separation stage to be discarded, collected as product, or further processed. In some embodiments, a pooled retentate stream is formed by combining each of the retentate streams from the plurality of membranes, where the pooled retentate stream is configured to exit the membrane separation stage to be discarded, collected as product, or further processed. A plurality of control valves may be provided upstream of each membrane in the plurality of membranes, where the control valves are configured to move between an open position and a closed position. A process measuring device may be provided in one or more of the inlet feed line, the pooled permeate stream, and/or the pooled retentate stream. The process measuring device is configured to acquire one or more measured value. The apparatus further includes a controller in electrical communication with the plurality of control valves and the process measuring device. The controller may include software stored therein and may be programmed to acquire one or more measured value in the inlet feed line, the pooled permeate stream, and/or the pooled retentate stream using the process measuring device, and adjust the total membrane surface area of the plurality of membranes by closing or opening one or more of the plurality of control valves in response to the measured value.

These and other advantages and features of the present invention will become more apparent from the following detailed description of the preferred embodiments of the present invention when viewed in conjunction with the accompanying drawings.

DESCRIPTION OF THE DRAWINGS

FIG. **1** is a block diagram of an example apparatus for separating a gas mixture in accordance with embodiments of the present disclosure.

FIG. **2** is a block diagram of the example apparatus shown in FIG. **1** used in conjunction with a process control system in accordance with embodiments of the present disclosure.

FIG. **3** is a block diagram of an example of a plurality of membranes configured within one or more of the first membrane stage, the second membrane stage, and/or the third membrane stage in accordance with embodiments of the present disclosure.

FIG. **4** is a block diagram of an alternative arrangement of the apparatus of FIG. **1** for separating a gas mixture in accordance with embodiments of the present disclosure.

FIG. **5** is a block diagram of an alternative arrangement of the apparatus of FIG. **1** for separating a gas mixture in accordance with embodiments of the present disclosure.

DETAILED DESCRIPTION

Before any embodiments of the disclosure are explained in detail, it is to be understood that the disclosure is not limited in its application to the details of construction and the arrangement of components set forth in the following description or illustrated in the following drawings. The disclosure is capable of other embodiments and of being practiced or of being carried out in various ways. Also, it is to be understood that the phraseology and terminology used herein is for the purpose of description and should not be regarded as limiting. The use of "including," "comprising," or "having" and variations thereof herein is meant to encompass the items listed thereafter and equivalents thereof as well as additional items. Unless specified or limited otherwise, the terms "mounted," "connected," "supported," and "coupled" and variations thereof are used broadly and encompass both direct and indirect mountings, connections, supports, and couplings. Further, "connected" and "coupled" are not restricted to physical or mechanical connections or couplings.

The following discussion is presented to enable a person skilled in the art to make and use embodiments of the disclosure. Various modifications to the illustrated embodiments will be readily apparent to those skilled in the art, and the generic principles herein can be applied to other embodiments and applications without departing from embodiments of the disclosure. Thus, embodiments of the disclosure are not intended to be limited to embodiments shown, but are to be accorded the widest scope consistent with the principles and features disclosed herein. The following detailed description is to be read with reference to the figures, in which like elements in different figures have like reference numerals. The figures, which are not necessarily to scale, depict selected embodiments and are not intended to limit the scope of embodiments of the disclosure. Skilled artisans will recognize the examples provided herein have many useful alternatives and fall within the scope of embodiments of the disclosure.

Referring now to FIG. **1**, an apparatus **10** for separating a gas mixture is shown. In some embodiments, the apparatus **10** includes at least a first membrane separation stage 1, a second membrane stage 2, and a third membrane stage 3. The first membrane separation stage 1 is provided in fluid communication with a gas source **11** comprising a reservoir of the gas mixture. In some embodiments, the gas source **11** may include biogas comprising a variety of components (e.g., methane, carbon dioxide, etc.). The biogas may be produced from sources such as agricultural waste, manure, municipal waste, plant material, sewage, green waste, food waste, anaerobic digester, or combinations thereof, and transferred to the apparatus **10** for separation.

In some embodiments, the apparatus **10** includes a first gas transport device **14** having a suction side connected to an inlet gas line **12** and a discharge side connected to a feed gas line **16**. The feed gas line **16** places the first gas transport device **14** in fluid communication with the first membrane stage 1, and the inlet gas line **12** places the first gas transport device **14** in fluid communication with the gas source **11**. In general, the gas transport device **14** may be provided as a mechanical device that adjusts (e.g., increases) the pressure of the gas mixture and may comprise a compressor, a blower or the like. In some embodiments, the first gas transport device **14** is configured to feed the gas mixture to the first separation stage 1 at a pressure that ranges between 1.5 bar (150 kPa) to 100 bar (10000 kPa), or more. In other embodiments, the first pressure may range between 5 bar (500 kPa) to 40 bar (4000 kPa), or 5 bar (500 kPa) to 35 bar (3500 kPa), or 5 bar (500 kPa) to 25 bar (2500 kPa). In other embodiments, the first pressure may range between about 1.5 bar (150 kPa) to about 100 bar (10000 kPa), or about 5 bar (500 kPa) to about 40 bar (4000 kPa), or about 5 bar (500 kPa) to about 35 bar (3500 kPa), or about 5 bar (500 kPa) to about 25 bar (2500 kPa).

The first membrane separation stage 1 includes one or more gas separation membranes configured to separate the gas mixture into a first retentate stream **18** and a first permeate stream **20**. The first permeate stream **20** includes gas components that pass through the gas separation membrane, while the first retentate stream **18** includes gas components retained by the gas separation membrane. In some embodiments, the gas separation membranes comprise a polymeric membrane. In some embodiments, suitable gas separation materials for use in the first membrane separation stage 1 include those that are selective for carbon dioxide over methane. A measure of the ability of a membrane to separate two gases is the selectivity, α, defined as the ratio of the gas permeabilities, P1/P2. Selectivity can also be expressed as:

$$\alpha_{12} = \frac{P_1}{P_2} = \left[\frac{D_1}{D_2}\right]\left[\frac{k_1}{k_2}\right]$$

where D is the diffusion coefficient of the gas in the membrane [cm2/s], which is a measure of the gas mobility, and k is the Henry's law sorption coefficient, which links the concentration of the gas in the membrane material to the pressure in the adjacent gas [cm^3 (STP)/cm^3·cmHg]. The ratio D1/D2 is the ratio of the diffusion coefficients of the two gases and can be viewed as the mobility selectivity, reflecting the different sizes of the two molecules. The ratio k_1/k_2 is the ratio of the Henry's law sorption coefficients of the two gases and can be viewed as the solubility selectivity, reflecting the relative condensabilities of the two gases. Depending on the nature of the polymer, either the diffusion or the sorption component of the permeability may dominate.

In some embodiments, the gas separation membrane of the first membrane separation stage 1 comprises a selectivity of CO_2/CH_4 that is greater than 1. In some embodiments the gas separation membrane of first membrane separation stage 1 comprises a selectivity of CO_2/CH_4 that is between 1 to 200. In some non-limiting examples, the first membrane separation stage 1 comprises a selectivity of CO_2/CH_4 that ranges between 3 to 85, or 10 to 80, or 20 to 70, or 30 to 60, or 45 to 55. In some non-limiting examples, the first membrane separation stage 1 comprises a selectivity of CO_2/CH_4 that ranges between about 1 to about 200, or about 3 to about 85, or about 10 to about 80, or about 20 to about 70, or about 30 to about 60, or about 45 to about 55.

In some embodiments, the gas separation membrane of the first membrane separation stage 1 comprises a selectivity of CO_2/CH_4 that is at least 5, at least 10, at least 15, at least 20, at least 25, at least 30, at least 35, at least 40, at least 45, at least 50, at least 55, at least 60, at least 65, at least 70, at least 80, at least 85, at least 90, at least 95, at least 100, at least 110, at least 120, at least 130, at least 140, at least 150, at least 160, at least 170, at least 180, at least 190, or at least 200. In some embodiments, the gas separation membrane of the first membrane separation stage 1 comprises a selectivity of CO_2/CH_4 that is at least about 5, at least about 10, at least about 15, at least about 20, at least about 25, at least about 30, at least about 35, at least about 40, at least about 45, at least about 50, at least about 55, at least about 60, at least about 65, at least about 70, at least about 80, at least about 85, at least about 90, at least about 95, at least about 100, at least about 110, at least about 120, at least about 130, at least about 140, at least about 150, at least about 160, at least about 170, at least about 180, at least about 190, or at least about 200.

Referring back to FIG. **1**, the first permeate stream **20** may be connected to a feed side of the second membrane separation stage 2. The second membrane separation stage 2 includes one or more gas separation membranes that separates the first permeate stream **20** into a second retentate stream **22** and a second permeate stream **24**. The second permeate stream **24** may be removed as a product, further processed, or discarded. The second retentate stream **22** is connected to a suction side of an optional second gas transport device **26** that is configured to adjust (e.g., increase) the pressure of the second retentate stream **22**. The optional second gas transport device **26** includes a discharge side that dispenses the gas components in the second retentate stream **22** into a continuing stream **28**, which places the optional second gas transport device **26** in fluid communication with the first retentate stream **18**. During operation, the second retentate stream **22** is mixed with the first retentate **18** through continuing stream **28** to form a mixed gas stream **30**.

The optional second gas transport device **26** may comprise a mechanical device that increases the pressure of the second retentate stream **22** and may comprise, for example, a compressor or a blower. In some embodiments, the optional second gas transport device **26** is configured to compress the second retentate stream **22** to a second pressure, which may be the same, or substantially the same, as the pressure of the first retentate stream **18**. In an alternative arrangement, the optional second gas transport device **26** may be configured in the first permeate stream **20**, and may be configured to compress the first permeate stream **20** such that the second pressure of the continuing stream **28** is the same, or substantially the same, as the pressure of the first retentate stream **18**.

In some embodiments, the first gas transport device **14** is designed to have a larger power output than the optional second gas transport device **26**. For example, the first gas transport device **14** and the optional second gas transport device **26** may be sized such that the power requirement (actual or theoretical) of the first gas transport device **14** to the optional second gas transport device **26** can range from at least 200:1 to at least 10:1, or from at least about 200:1 to at least about 10:1. In some embodiments, the power requirement of the first gas transport device **14** to the optional second gas transport device **26** is at least 10:1, at least 15:1, at least 20:1, at least 25:1, at least 30:1, at least 35:1, at least 40:1, at least 45:1, at least 50:1, at least 55:1, at least 60:1, at least 65:1, at least 70:1, at least 75:1, at least 80:1, at least 90:1, at least 95:1, or 100:1. In some embodiments, the power requirement of the first gas transport device **14** to the optional second gas transport device **26** is at least about 10:1, at least about 15:1, at least about 20:1, at least about 25:1, at least about 30:1, at least about 35:1, at least about 40:1, at least about 45:1, at least about 50:1, at least about 55:1, at least about 60:1, at least about 65:1, at least about 70:1, at least about 75:1, at least about 80:1, at least about 90:1, at least about 95:1, or about 100:1.

In some non-limiting examples, the gas mixture comprises biogas and, during operation, the methane content in the mixed gas stream **30** is at least 1.1 times greater than the methane content in the feed gas line **16**, or at least about 1.1 times greater than the methane content in the feed gas line **16**. In some embodiments, the methane content in the mixed gas stream **30** is at least 1.2 times greater, at least 1.3 times greater, at least 1.4 times greater, at least 1.5 times greater, at least 1.6 times greater, at least 1.7 times greater, at least 1.8 times greater, at least 1.9 times greater, or at least 2 times greater than the methane content in the feed gas line **16**. In some embodiments, the methane content in the mixed gas stream **30** is at least about 1.2 times greater, at least about 1.3 times greater, at least about 1.4 times greater, at least about 1.5 times greater, at least about 1.6 times greater, at least about 1.7 times greater, at least about 1.8 times greater, at least about 1.9 times greater, or at least about 2 times greater than the methane content in the feed gas line **16**.

In some embodiments, the gas separation membrane of the second membrane separation stage 2 comprises a polymeric membrane. In some embodiments, the gas separation membrane of the second membrane separation stage 2 may comprise a selectivity that is the same or different than the first membrane separation stage 1. In some embodiments, the gas separation membrane of the second membrane separation stage 2 comprises a selectivity of CO_2/CH_4 that is greater than 1. In some embodiments the gas separation membrane of second membrane separation stage 2 comprises a selectivity of CO_2/CH_4 that is 1 to 200, or about 1 to about 200. In some non-limiting examples, the second membrane separation stage 2 comprises a selectivity of CO_2/CH_4 that ranges between 3 to 85, or 10 to 80, or 20 to 70, or 30 to 60, or 45 to 55. In some non-limiting examples, the second membrane separation stage 2 comprises a selectivity of CO_2/CH_4 that ranges between about 3 to about 85, or about 10 to about 80, or about 20 to about 70, or about 30 to about 60, or about 45 to about 55.

In some embodiments, the gas separation membrane of the second membrane separation stage 2 comprises a selectivity of CO_2/CH_4 that is at least 5, at least 10, at least 15, at least 20, at least 25, at least 30, at least 35, at least 40, at least 45, at least 50, at least 55, at least 60, at least 65, at least 70, at least 80, at least 85, at least 90, at least 95, at least 100, at least 110, at least 120, at least 130, at least 140, at least 150, at least 160, at least 170, at least 180, at least 190, or at least 200. In some embodiments, the gas separation membrane of the second membrane separation stage 2 comprises a selectivity of CO_2/CH_4 that is at least about 5, at least about 10, at least about 15, at least about 20, at least about 25, at least about 30, at least about 35, at least about 40, at least about 45, at least about 50, at least about 55, at least about 60, at least about 65, at least about 70, at least about 80, at least about 85, at least about 90, at least about 95, at least about 100, at least about 110, at least about 120, at least about 130, at least about 140, at least about 150, at least about 160, at least about 170, at least about 180, at least about 190, or at least about 200.

Referring back to FIG. **1**, the mixed gas stream **30** is connected to a feed side of the third membrane separation stage 3. The third membrane separation stage 3 includes a gas separation membrane that separates the mixed gas stream **30** into a third retentate stream **32** and a third permeate stream **34**. The third retentate stream **32** may be withdrawn as a product, further processed, or discarded. In some embodiments, the third permeate stream **34** is recycled back to the inlet gas line **12**. Alternatively, or additionally, the third permeate stream **34** may be recycled via a recycle line **35** (optional) back to the gas source **11**, for example, to a feed inlet **8** configured upstream of the gas source **11**. Alternatively, the third permeate stream **34** may be further processed, or discarded.

In some embodiments, the gas separation membrane of the third membrane separation stage 3 comprises a polymeric membrane. In some embodiments, the gas separation membrane of the third membrane separation stage 3 may comprise a selectivity that is the same or different than the first membrane separation stage 1 and/or the second membrane separation stage 2. In some embodiments, the gas separation membrane of the second membrane separation stage 2 comprises a selectivity of CO_2/CH_4 that is greater than 1, or greater than about 1. In some embodiments the gas separation membrane of third membrane separation stage 3 comprises a selectivity of CO_2/CH_4 that is 1 to 200, or about 1 to about 200. In some non-limiting examples, the third membrane separation stage 2 comprises a selectivity of CO_2/CH_4 that ranges between 3 to 85, or 10 to 80, or 20 to 70, or 30 to 60, or 45 to 55. In some non-limiting examples, the third membrane separation stage 2 comprises a selectivity of CO_2/CH_4 that ranges between about 3 to about 85, or about 10 to about 80, or about 20 to about 70, or about 30 to about 60, or about 45 to about 55.

In some embodiments, the gas separation membrane of the third membrane separation stage 3 comprises a selectivity of CO_2/CH_4 that is at least 5, at least 10, at least 15, at least 20, at least 25, at least 30, at least 35, at least 40, at least 45, at least 50, at least 55, at least 60, at least 65, at least 70, at least 80, at least 85, at least 90, at least 95, at least 100, at least 110, at least 120, at least 130, at least 140, at least 150, at least 160, at least 170, at least 180, at least 190, or at least 200. In some embodiments, the gas separation membrane of the third membrane separation stage 3 comprises a selectivity of CO_2/CH_4 that is at least about 5, at least about 10, at least about 15, at least about 20, at least about 25, at least about 30, at least about 35, at least about 40, at least about 45, at least about 50, at least about 55, at least about 60, at least about 65, at least about 70, at least about 80, at least about 85, at least about 90, at least about 95, at least about 100, at least about 110, at least about 120, at least about 130, at least about 140, at least about 150, at least about 160, at least about 170, at least about 180, at least about 190, or at least about 200.

In some embodiments, the first membrane separation stage 1, the second membrane separation stage 2, and/or the third membrane separation stage 3 may include one or more vacuum pumps arranged downstream of the respective membrane separation stage to assist in producing a pressure differential across the gas separation membrane. In some embodiments, the second membrane separation stage 2 and the third membrane separation stage 3 may include one or more vacuum pumps arranged downstream of the respective membrane separation stage to assist in producing a pressure differential across the gas separation membrane. Additionally, or alternatively, the first membrane separation stage 1, the second membrane separation stage 2, and the third membrane separation stage 3 may optionally include a sweep gas flowing along the permeate side of each respective stage to assist the separation.

In some embodiments, a pretreatment unit (not shown in FIG. **1**) is configured in the inlet gas line **12** and may be optionally used to remove contaminants from the gas mixture. For example, the pretreatment unit may be configured to perform filtration (e.g., by activated carbon), adsorption, absorption, or condensation to remove one or more contaminants from the gas mixture including, but not limited to siloxanes, volatile organic compounds, H_2S, water vapor, and combinations thereof.

The fluid communication within the apparatus **10** disclosed in FIG. **1** offers several advantages over conventional system. For example, one advantage of recycling the second retentate stream **22** to the first retentate stream **18** is that it allows for a reduction in the total amount of gas that is recycled back the first gas transport device **14**, as opposed to recycling the second retentate stream **22** back to the inlet gas line **12**. This allows for the first gas transport device **14** to be smaller, thereby reducing initial investment costs, lowering operation costs, and maximizing the total system efficiency to allow for a reduction in total membrane area as compared to conventional systems. The apparatus **10** additionally offers improved process control, as will be described.

FIG. **2** illustrates the apparatus **10** used in conjunction with an example process control system according to some embodiments of the present disclosure. The process control system may be configured to control the operation of the apparatus **10**. For example, the process control system may assist in starting the process, stopping the process, and adjusting process parameters to change the process performance. In some embodiments, the control system may be designed to control the concentration of a desired component in the third retentate stream **32** or the second permeate stream **24**, and/or to monitor and adjust system parameters to control energy consumption to reduce operational costs.

In some embodiments the control system may adjust or control a multitude of process parameters to maintain a product gas purity. For example, the product gas purity leaving the third retentate stream **32** may be controlled using the control system to be at least 80%, at least 85%, at least 90%, at least 91%, at least 92%, at least 93%, at least 94%, at least 95%, at least 96%, at least 97%, at least 98%, at least 99%, at least 99.5%, or at least 99.9%. In some embodiments, the product gas purity leaving the third retentate stream **32** may be controlled using the control system to be at least about 80%, at least about 85%, at least about 90%, at least about 91%, at least about 92%, at least about 93%, at least about 94%, at least about 95%, at least about 96%, at least about 97%, at least about 98%, at least about 99%, at least about 99.5%, or at least about 99.9%. In some non-limiting examples, the product gas in the third retentate stream **32** comprises methane.

In some embodiments, the process control system includes a controller **36**, one or more process measuring devices (e.g., **38***a*-**38***h*), one or more process control devices (e.g., the first gas transport device **14**, the optional second gas transport device **26**, one or more valves **40***a*-**40***e*, and temperature control devices **42***a*-**42***c*), and suitable connections that allows process information acquired by the one or more measuring devices to be transferred to the controller **36**, and output information from the controller **36** to be transferred to the one or more process control devices to perform a process control action. Example process control actions may include using the process control devices to alter one or more of a pressure, flowrate, and/or temperature of a process stream based on one or more measured values from the process measuring devices.

Suitable connections may include transmitters that allow process signals, such as electrical signals or gas pressure signals (e.g., air, nitrogen, etc.), to be transmitted between the controller **36** and the measuring devices (e.g., **38***a*-**38***h*) and the process control devices (e.g., **14**, **26**, **40***a*-**40***d*, and **42***a*-**42***c*). In some aspects, the electrical signals may be transferred via a wired connection or through a wireless network connection. Other hardware elements may be included in the process control system, for example, transducers, analog-to-digital (A/D) converters, and digital-to-analog (D/A) converters that allow process information to be recognizable in computer form, and computer commands accessible to the process. For visual clarity, the connections between the controller **36**, the one or more measuring devices, and the one or more process control devices have been omitted from FIG. **2**.

In some embodiments, the apparatus **10** includes one or more process measuring devices (e.g., **38***a*-**38***h*). For example, the apparatus **10** may include one or more of an inlet gas line measuring device **38***a* provided in the inlet gas line **12**, a feed gas line measuring device **38***b* configured in the gas feed line **16**, a first permeate measuring device **38***c* configured in the first permeate stream **20**, a first retentate measuring device **38***d* configured in the first retentate stream **18**, a second permeate measuring device **38***e* configured in the second permeate stream **24**, a second retentate measuring device **38***f* configured in the second retentate stream **28**, a third permeate measuring device **38***g* configured in the third permeate stream **34**, and a third retentate measuring device **38***h* configured in the third retentate stream **32**. Each of the process measuring devices **38***a*-**38***h* may be configured to measure a process parameter, or multiple process parameters, such as pressure, temperature, flow, composition, viscosity, humidity, moisture, or density.

In some embodiments, the one or more measuring devices **38***a*-**38***h* includes a pressure measuring device. Suitable pressure measuring devices include, but are not limited to, pressure transducers, diaphragm bellows, Bourdon tubes, strain gauge, piezoelectric sensors, ionization gauges, and combinations thereof.

In some embodiments, the one or more measuring devices **38***a*-**38***h* includes a flow measuring device. Suitable flow measuring devices include, but are not limited to, pitot tubes, orifice flow meters, turbine flow meters, electromagnetic field flow meters, neutron bombardment flow meters, ultrasound flow meters, hot-wire anemometry flow meters, and angular momentum flow meters, Coriolis mass flow meters, and combinations thereof.

In some embodiments, the one or more measuring devices **38***a*-**38***h* includes a temperature measuring device. Suitable temperature measuring devices include, but are not limited to, thermocouples, thermistors (e.g., resistance sensor), oscillating quartz crystal thermometer, radiation pyrometers, and combinations thereof.

In some embodiments, the one or more measuring devices **38***a*-**38***h* includes a density measuring device. Suitable density measuring devices include, but are not limited to, vibrating tube densometers, X-ray densometers, differential pressure densometers, and combinations thereof.

In some embodiments, the one or more measuring devices **38***a*-**38***h* includes a viscosity measuring device. Suitable viscosity measuring devices include, but are not limited to, capillary viscometers, vibrating ball viscometers, and combinations thereof.

In some embodiments, the one or more measuring devices **38***a*-**38***h* includes a humidity measuring device. Suitable humidity measuring devices include hygrometers that may be configured, for example, to measure precise dew point measurements.

In some embodiments, the one or more measuring devices **38***a*-**38***h* includes a composition measuring device. Suitable composition measuring devices include, but are not limited to, potentiometry sensors, moisture content sensors (hygrometry or psychrometry), gas chromatography, refractive index device, ultrasound, spectroscopy system (e.g., UV, visible, IR, Mossbauer, Raman, atomic-emission device, X-ray device, electron, ion, nuclear magnetic resonance), polarography device, conductimetry device, mass spectrometry system, differential thermal analysis device, and thermogravi-metric analysis system, and combinations thereof.

Referring back to FIG. **2**, the apparatus **10** includes one or more process control devices, such as valves (e.g., **40***a*-**40***e*), temperature control devices (e.g., **42***a*-**42***c*), the first gas transport device **14**, and the second gas transport device **26**. In some embodiments, a third gas transport device (not shown), such as a compressor or a blower, may be provided in the first permeate stream **20**.

In some embodiments, the apparatus **10** includes one or more valves **40***a*-**40***f* that is adjustable between a fully-open position and a fully-closed position, and is used to regulate the pressure of the process streams. For example, the apparatus **10** may include a first permeate valve **40***a* provided in the first permeate stream **20**, a first retentate valve **40***b*, a second permeate valve **40***c* provided in the permeate stream **24**, a third permeate valve **40***d* provided in the third permeate stream **34**, a third retentate valve **40***e* provided in the third retentate stream **32**, and a second retentate valve **40***f* provided in the continuing stream **28**. Example valves for use in apparatus **10** include, without limitation, motor-driven valves, pneumatic values, and manually adjustable valves.

The apparatus **10** may include one or more temperature control devices (e.g., **42***a*-**42***c*) that may be used to adjust the temperature (e.g., heat or cool) of the process streams. For example, the apparatus **10** may include one or more of a first stage temperature control device **42***a* configured in the gas feed line **16**, a second stage temperature control device **42***b* configured in the first permeate stream **20**, and a third stage temperature control device **42***c* configured in the second retentate stream **28**. Alternatively, the third stage temperature control device **42***c* may be configured in the first retentate stream **18** or the mixed gas stream **30**. Suitable temperature control devices **42***a*-**42***c* include devices that heat or cool the process stream, such as heat exchangers or electric heaters.

The controller **36** includes a processor **44** and a memory **48** that includes software **50** and data **52**, and is designed for storage and retrieval of processed information to be processed by the processor **44**. The processor **44** includes an input **54** that is configured to receive process signals from the one or more of the measuring devices (**38***a*-**38***h*) and the one or more of the process control devices (e.g., **14**, **26**, **40***a*-**40***f*, and **42***a*-**42***c*) via the input **54**. The controller **36** may operate autonomously or semi-autonomously, or may read executable software instructions from the memory **48** or a computer-readable medium (e.g., a hard drive, a CD-ROM, flash memory), or may receive instructions via the input **54** from a user, or another source logically connected to a computer or device, such as another networked computer or server. For example, the server may be used to control the apparatus **10** via the controller **36** on-site or remotely.

The processor **44** may process the process the signals to generate an output **56**, which may take the form of a process control action. Example process control actions may include sending signals to the one or more process control devices (e.g., **14**, **26**, **40***a*-**40***f*, and **42***a*-**42***c*) to effectuate a change in one or more process parameters (e.g., pressure, flowrate, and/or temperature) of one or more process streams in apparatus **10**.

In some embodiments, the controller **36** includes programming to adjust the process parameters in the apparatus **10** using the one or more process control devices (e.g., **14**, **26**, **40***a*-**40***f*, and **42***a*-**42***c*) in response to measured values obtained from the one or more measuring devices (e.g., **38***a*-**38***h*) to maintain a desired set-point in the third retentate stream **32**, such as a desired pressure, a desired component composition, and/or a desired flowrate.

As a first example, the controller **36** may be programmed to monitor one or more process parameters in the third retentate stream **32** (e.g., methane content, pressure, etc.) using the third retentate measuring device **38***h*. Once a change in the process parameter is detected or a user-specified threshold has been reached, the pressure of the third retentate stream **32** may be adjusted using the third retentate valve **40***e* in order to maintain the desired set-point. Alternatively or additionally, the feed gas line measuring device **38***b* may be used to monitor one or more process parameters in the feed gas line **16**, where the desired set-point in the third retentate stream **32** is maintained by steering the pressure in the third permeate stream **34** using the third permeate valve **40***d*.

In some embodiments, the controller **36** includes programming to adjust process parameters in the apparatus **10** using the one or more process control devices (e.g., **14**, **26**, **40***a*-**40***f*, and **42***a*-**42***c*) in response to measured values obtained from the one or more measuring devices (e.g., **38***a*-**38***h*) to maintain a desired set-point in the second permeate stream **24**, such as a desired pressure, a desired component composition, and/or a desired flowrate.

As a second example, the controller **36** may include software **50** that is programmed to acquire one or more process parameters in the second permeate stream **24** using the second permeate measuring device **38***e*. Once a change is detected or a user-specified threshold has been reached, the pressure of the second permeate stream **24** may be adjusted using the second permeate valve **40***c* to maintain the desired set-point.

In some embodiments, the controller **36** includes programming to adjust process parameters in the apparatus **10** using the one or more process control devices (e.g., **14**, **26**, **40***a*-**40***f*, and **42***a*-**42***c*) in response to measured values obtained from the one or more measuring devices (e.g., **38***a*-**38***h*) to maintain a desired set-point in the first permeate stream **20**, such as a desired pressure, a desired component composition, and/or a desired flowrate.

In a third example, the controller **36** is programmed to acquire one or more process parameters in the first permeate stream **20** using the first permeate measuring device **38***c*. Once a change is detected or a user-specified threshold has been reached, the pressure of the first permeate stream **20** may be adjusted using the first permeate valve **40***a* to maintain the desired set-point. In one non-limiting example, a gas component composition (e.g., methane) in the first permeate stream **20** is measured and controlled to maintain the desired set-point by adjusting the pressure in the first permeate stream **20** using the first permeate valve **40***a*.

In some embodiments, the controller **36** includes programming to adjust process parameters in the apparatus **10** using the one or more process control devices (e.g., **14**, **26**, **40***a*-**40***f*, and **42***a*-**42***c*) in response to measured values obtained from the one or more measuring devices (e.g., **38***a*-**38***h*) to maintain a desired set-point in the third permeate stream **34**, such as a desired pressure, a desired component composition, and/or a desired flowrate.

In a fourth example, the controller **36** is programmed to acquire one or more process parameters in the third permeate stream **34** using the third permeate measuring device **38***g*. Once a change is detected or a user-specified threshold has been reached, the pressure of the third permeate stream **34** may be adjusted using the third permeate valve **40***d* to obtain the desired set-point. In one non-limiting example, the pressure of the third permeate stream **34** is measured and controlled to be the same or substantially the same as the pressure of the inlet gas line **12**, which may be measured using the inlet gas line measuring device **38***a*.

In some embodiments, the controller **36** includes programming to adjust process parameters in the apparatus **10** using the one or more process control devices (e.g., **14**, **26**, **40***a*-**40***f*, and **42***a*-**42***c*) in response to measured values obtained from the one or more measuring devices (e.g., **38***a*-**38***h*) to maintain a desired set-point in the second retentate stream **22** and/or the continuing stream **28**, such as a desired pressure, a desired component composition, and/or a desired flowrate.

In a fifth example, the controller **36** is programmed to acquire one or more process parameters in the second retentate stream **22** and/or the continuing stream **28** using the second retentate measuring device **38***f*. Once a change is detected or a user-specified threshold has been reached, the pressure of the continuing stream **28** may be adjusted using the second retentate valve **40***f* to maintain the desired set-point in the continuing stream **28**. In one non-limiting example, the pressure of the continuing stream **28** is measured using the second retentate measuring device **38***f* and adjusted using the second retentate valve **40***f* such that the pressure of the continuing stream **28** is sufficient to be added to the first retentate stream **18**. For example, the continuing stream **28** may be adjusted by the second retentate valve **40***f* to be the same or substantially the same as the pressure of the first retentate **18**, which may be measured using the first retentate measuring device **38***d*.

Additionally, or alternatively, the controller **36** may be programmed to acquire one or more process parameter in the first retentate stream **18** using the first retentate measuring device **38***d*. Once a change is detected or a user-specified threshold has been reached, the pressure of the first retentate stream **18** may be adjusted using the first retentate valve **40***b*. In one non-limiting example, the pressure of the first retentate stream **18** is measured using the first retentate measuring device **38***d* and is adjusted using the first retentate valve **40***b* such that the pressure of the first retentate stream **18** is sufficient to be combined with the continuing stream **28**.

In some embodiments, the controller **36** includes programming to adjust process parameters in the apparatus **10** using the one or more process control devices (e.g., **14**, **26**, **40***a*-**40***f*, and **42***a*-**42***c*) in response to measured values obtained from the one or more measuring devices (e.g., **38***a*-**38***h*) to maintain a desired set-point in the inlet gas line **12** and/or the feed gas line **16**, such as a desired pressure, a desired component composition, and/or a desired flowrate.

In some embodiments, the controller **36** includes programming to adjust process parameters in the apparatus **10** using the one or more process control devices (e.g., **14**, **26**, **40***a*-**40***f*, and **42***a*-**42***c*) in response to measured values obtained from the one or more measuring devices (e.g., **38***a*-**38***h*) to maintain a desired temperature set-point for the first membrane separation stage 1, and/or the second membrane stage 2, and/or the third membrane separation stage 3.

In a sixth example, the controller **36** is programmed to acquire one or more process parameters in the apparatus **10** using one or more of the process measuring devices (e.g., **38***a*-**38***h*) and adjust the temperature of operation temperature of the first membrane separation stage 1, and/or the second membrane separation stage 2, and/or the third membrane separation stage 3 using the first temperature control device, and/or the second stage temperature control device **42***b*, and/or the third stage temperature control device **42***c* to obtain a desired temperature set-point. The temperature of the first membrane separation stage 1, the second membrane stage 2, the third membrane separation stage 3 may be controlled to be the same or different.

Referring now to FIG. **3**, one or more of the membrane stages 1-3 in apparatus **10** may include a plurality of gas separation membranes **60** configured therein. The plurality of gas separation membranes **60** may be configured in parallel or in series.

In some embodiments, the one or more of the membrane stages 1-3 are configured with a first gas separation membrane unit **60***a*, a second gas separation membrane unit **60***b*, and (optionally, as indicated by break lines **61**) one or more additional gas separation membrane units **60***n* in fluid communication with the incoming stream (e.g., **16**, **20**, or **30**). The incoming stream is then split into multiple inlet streams to feed into the plurality of gas separation membranes **60**. For example, the incoming stream may be split into a first interior membrane stage inlet stream **62***a*, a second interior membrane stage inlet stream **62***b*, and optionally one or more additional interior membrane stage inlet stream **62***n*. Each of the interior membrane stage inlet streams **62***a*, **62***b*, and **62***n* may be configured with a valve **64***a*, **64***b*, and **64***n* to regulate the flow rate to the respective gas separation membrane unit.

In some embodiments, each of the gas separation membrane units **60***a*, **60***b*, and **60***n* includes a gas separation membrane that separates the incoming gas mixture into interior membrane stage permeate streams **66***a*, **66***b*, **66***n* and retentate stream **68***a*, **68***b*, **68***n*, respectively. The permeate streams **66***a*, **66***b*, **66***n* may be combined to form a pooled permeate stream (e.g., **20**, **24**, and **34**) that exits the one or more membrane stages 1-3. In some embodiments, permeate control valves **70***a*, **70***b*, **70***n* may be configured in the permeate streams **66***a*, **66***b*, **66***n* to regulate the gas flow rate to the pooled permeate stream. In an alternative arrangement, the permeate control valves **70***a*, **70***b*, **70***n* may be combined into a single valve positioned in the pooled permeate stream (e.g., **20**, **24**, **34**). Similarly, each of the retentate streams **68***a*, **68***b*, **68***n* may be combined to form a pooled retentate stream (e.g., **18**, **22**, and **32**) that exits the one or more membrane stages 1-3. Retentate control valves **72***a*, **72***b*, **72***n* may be configured to regulate the gas flow rate of the pooled retentate stream. In an alternative arrangement, the retentate control valves **72***a*, **72***b*, **72***n* may be combined into a single valve positioned in the pooled retentate stream (e.g., **18**, **22**, **32**).

In some embodiments, the controller **36** is placed in electrical communication with the first interior control valve **64***a*, the second interior control valve **64***b*, the one or more additional interior control valve **64***n*, the permeate control valves **70***a*, **70***b*, **70***n*, and the retentate control valves **72***a*, **72***b*, **72***n*.

In some embodiments, the controller **36** includes programming to adjust the total surface area (e.g., increase or decrease) of the one or more membrane stages 1-3 using the one or more interior control devices (e.g., **64***a*, **64***b*, **64***n*, **70***a*, **70***b*, **70***n*, **72***a*, **72***b*, and **72***n*). The total surface area of the one or more membrane stages 1-3 may be adjusted manually by closing and opening the one or more interior control devices, for example, by a user. Alternatively or additionally, the total surface area of the one or more membrane stages 1-3 may be adjusted in response to measured values obtained from the one or more measuring devices (e.g., **38***a*-**38***h*) to maintain a desired set-point in one or more of the process streams, such as a desired pressure, a desired component composition, and/or a desired flowrate. In some embodiments, the total surface area of the membrane stages 1-3 have a surface area ratio of 1:1:1, or about 1:1:1, or 1:2:1, or about 1:2:1, or from 1:1:1 to 1:4:1, or from about 1:1:1 to about 1:4:1. Tests have shown that adjusting the surface area ratio of the first membrane separation stage 1, the second membrane stage 2, and the third membrane separation stage 3 may enable, for example, the use of smaller or larger capacity compressors to be used in conjunction with apparatus **10**, to adapt the apparatus **10** to specific needs.

As a seventh example, the controller **36** is programmed to acquire one or more process parameters in the apparatus **10** using the one or more of the process measuring devices (e.g., **38***a*-**38***h*) and adjust the total surface area of the one or more membrane separation stages 1-3 by closing (e.g., to decrease surface area) or opening (e.g., to increase surface area) the valves to the first gas separation membrane unit **60***a*, the second gas separation membrane unit **60***b*, and/or the one or more additional gas separation membrane units **60***n*.

For example, if the flowrate of the gas mixture to apparatus **10** decreases, which may be measured using the inlet gas line measuring device **38***a* or the feed gas line measuring device **38***b*, it may be advantageous to reduce the total surface area of the one or more membrane stages 1-3 to maximize efficiency and to maintain a desired set-points during operation. This may be done, for example, by closing the one or more additional inlet cascade valve **64***n*, the one or more additional permeate valve **70***n*, and the one or more additional retentate valve **72***n* in response to the change in flowrate of the gas mixture to the apparatus **10**.

Alternatively, if the flowrate of the gas mixture to the apparatus **10** increases, it may be advantageous to increase the total surface area of the one or more membrane stages 1-3. This may be done, for example, by opening the one or more additional inlet interior valve **64***n*, the one or more additional permeate valve **70***n*, and the one or more additional retentate valve **72***n* in response to the change in flowrate of the gas mixture to the apparatus **10**.

FIG. **4** depicts an alternative arrangement of apparatus **10***a* according to some embodiments of the present disclosure. The apparatus **10***a* includes similar features as described with respect to FIGS. **1**-**3**, and further includes a second continuing stream **37** that places the third retentate stream **32** in fluid communication with the continuing stream **28**. During operation, the third retentate stream **32** is mixed with the second continuing stream **37** to form a mixed retentate stream **39**. The mixed retentate stream **39** may be withdrawn as product, discarded, or further processed. In some embodiments, the optional second gas transport device **26** is removed from apparatus **10***a*. In some embodiments a measuring device (not pictured) and a valve (not pictured) are provided in the mixed retentate stream **39**.

In some embodiments, the apparatus **10***a* further includes one or more process control devices, such as a valve **40***f*, that controls the pressure and gas flowrate of the continuing stream **28** and the second continuing stream **37**. In some embodiments, the valve **40***f* may be adjusted between a fully-open position and a fully-closed position, which may be used to close the gas flow to the second continuing stream **37** and direct all the flow to the continuing stream **28**, or vice versa. Alternatively, the valve **40***f* may be adjustable to divert a fraction of the gas to the second continuing stream **37** and the remaining fraction to the continuing stream **28**. The valve **40***f* may be, for example, a motor-driven valve, a pneumatic valve, or a manually adjustable valve.

In some embodiments, the apparatus **10***a* includes one or more additional process measuring devices, such as a second continuing stream measuring device **38***i*. The second continuing stream measuring device **38***i* may be configured to measure a process parameter, or multiple process parameters, such as pressure, temperature, flow, composition, viscosity, humidity, moisture, or density. The second continuing stream measuring device **38***i* may include one or more of the process measuring devices described above.

In some embodiments, the controller **36** is configured to be in electrical communication with the valve **40***f* and the second continuing stream measuring device **38***i*. The controller **36** may include programming to adjust process parameters in apparatus **10***a* using the one or more process control devices (e.g., **14**, **26**, **40***a*-**40***f*, and **42***a*-**42***c*) in response to measured values obtained from the one or more process measuring devices (e.g., **38***a*-**38***i*) to maintain a desired set-point in the one or more process stream in apparatus **10***b*, such as a desired pressure, a desired component composition, and/or a desired flowrate.

In an eighth example, the controller **36** may be programmed to acquire one or more process parameters in the second continuing stream **37** using the second continuing stream measuring device **38***i*. Once a change is detected or a user-specified threshold has been reached, the pressure or flow rate of the second continuing stream **37** and the continuing stream **28** may be adjusted using the valve **40***f*. In one non-limiting example, the pressure or flowrate of the second continuing stream **37** is measured using the second continuing stream measuring device **38***i* and adjusted such that the pressure of the second continuing stream **37** is sufficient to be added to the third retentate stream **32**.

FIG. **5** depicts an alternative arrangement of apparatus **10***b* according to some embodiments of the present disclosure. The apparatus **10***b* includes similar features as described with respect to FIGS. **1**-**4**, and further includes a permeate sweep stream **41** that places the second continuing stream **37** in fluid communication with the permeate side of the third membrane separation stage 3. In some embodiments, the permeate sweep stream **41** is configured to enter the permeate side of the third membrane separation stage 3 such that the gas is swept across the membrane, and subsequently discharges into the third permeate stream **34**. In some embodiments, the optional second gas transport device **26** is not present in apparatus **10***b*. In some embodiments a measuring device (not pictured) and a valve (not pictured) are provided in the mixed retentate stream **39**.

In some embodiments, the apparatus **10***b* further includes one or more process control devices, such as a valve **40***g*, that controls the pressure and gas flowrate of the second continuing stream **37** and the permeate sweep stream **41**. In some embodiments, the valve **40***g* may be adjusted between a fully-open position and a fully-closed position, which may be used to close the gas flow to the permeate sweep stream **41** and direct all the flow to the second continuing stream **37**, or vice versa. Alternatively, the valve **40***g* may be adjustable to divert a fraction of the gas to the permeate sweep stream **41** and the remaining fraction to the second continuing stream **37**. The valve **40***g* may be, for example, a motor-driven valve, a pneumatic valve, or a manually adjustable valve.

In some embodiments, the apparatus **10***b* includes one or more additional process measuring devices, such as a sweep flow measuring device **38***j*. The sweep flow stream measuring device **38** may be configured to measure a process parameter, or multiple process parameters, such as pressure, temperature, flow, composition, viscosity, humidity, moisture, or density. The sweep flow measuring device **38***j* may include one or more of the process measuring devices described above.

In some embodiments, the controller **36** is configured to be in electrical communication with the valve **40***g* and the sweep flow measuring device **38***j*. The controller **36** may include programming to adjust process parameters in apparatus **10***b* using the one or more process control devices (e.g., **14**, **26**, **40***a*-**40***g*, and **42***a*-**42***c*) in response to measured values obtained from the one or more process measuring devices (e.g., **38***a*-**38***j*) to maintain a desired set-point in the one or more process stream in apparatus **10***b*, such as a desired pressure, a desired component composition, and/or a desired flowrate.

In a ninth example, the controller is programmed to acquire one or more process parameters in the permeate sweep stream **41** and the third retentate stream **32** using the sweep flow measuring device **38***j* and third retentate measuring device **38***h*, respectively. Once a change is detected or a user-specified threshold has been reached in the third retentate measuring device **38***h*, the pressure or flow rate in the permeate sweep flow stream may be adjusted using the valve **40***g* to obtain a desired set-point in the third retentate stream **32**.

EXAMPLES

The following examples set forth, in detail, ways in which the apparatus **10** separates a gas mixture, and will enable one of skill in the art to more readily understand the principles thereof. The following examples are presented by way of illustration and are not meant to be limiting in any way.

Example 1

Table 1 illustrates the Example 1 simulation data, which illustrates an example process of separating methane from biogas using apparatus **10**, as depicted in FIGS. **1** and **2**. For the simulation, the biogas from the gas source **11** was simulated having 68% carbon dioxide (w/w), 30.99% methane (w/w), 0.22% oxygen (w/w), and 0.79% nitrogen (w/w).

The results of the simulation are shown in Table 1:

TABLE 1

	Drawing Reference Number				
	12	16	18	30	32
Stream Name	Inlet Gas Line	Feed Gas Line	Stage 1 Ret	Stage3 Feed	Stage3 Ret
Temp (K)	298.15	298.15	298.05	299.56	299.53
Temp (° C.)	25.0	25.0	24.9	26.41	26.38
Pres (kPa)	105	1130	1114	1114	1107
Vapor mole fraction	1.00	1.00	1.00	1.00	1.00
Std vap 0 C. m3/h	1000.00	1409.09	817.74	970.31	561.22
Flow rates in kg/h					
Methane	393.67	470.22	429.88	468.08	391.53
Carbon Dioxide	863.96	1444.88	402.94	592.22	11.29
Nitrogen	10.00	13.90	11.79	13.69	9.78
Component mass %					
Methane	30.99	24.28	50.68	43.36	94.69
Carbon Dioxide	68.00	74.62	47.51	54.86	2.73
Nitrogen	0.79	0.72	1.39	1.27	2.37

As shown in Table 1, the third retentate stream **32** is significantly higher in methane gas purity (94.69%) compared to methane gas purity (30.99%) in the initial inlet gas line **12**. The fact that the apparatus **10** is able to separate methane from biogas, to a purity of greater than 80%, and further greater than 94%, suggests that the apparatus **10** is suitable commercial gas separation, and, in particular, for commercial methane gas separation from biogas.

Example 2

Sample A. Sample A (S-A) is run using apparatus **10**, as depicted in FIGS. **1** and **2**. The first gas transport device **14** and the second gas transport device **26** are each compressors. In other words, the apparatus is equipped with two compressors.

Comparative Sample B. Comparative Sample B (CS-B) is run using Comparative Apparatus 3T. The Comparative Apparatus 3T is the same as the apparatus **10** depicted in FIGS. **1** and **2**, except that the second retentate stream is immediately recycled back to the initial gas inlet via a recycle line, instead of forming a mixed gas stream that flows through the third membrane separation stage. Comparative Apparatus 3T utilizes a single compressor in the location of the first gas transport device **14** depicted in FIG. **1**.

Comparative Sample C. Comparative Sample C (CS-C) is run using Comparative Apparatus 3TI. The Comparative Apparatus 3TI is the same as the apparatus Comparative Apparatus 3T descriptive above, except that a booster device is connected to the line carrying the first permeate stream from the first membrane separation stage to the second membrane separation stage. As in Comparative Apparatus 3T, the Comparative Apparatus 3TI second retentate stream is immediately recycled back to the initial gas inlet via a recycle line, instead of forming a mixed gas stream that flows through the third membrane separation stage.

The first membrane stage, the second membrane stage, and the third membrane stage of each of Sample A, Comparative Sample B, and Comparative Sample C are equipped with an "FH" membrane (commercially available as COxxxFH, from Ube Industries, Ltd., where "xxx" refers to the size of the membrane), as shown in Table 2.

The inlet gas line **12** of each of Sample A, Comparative Sample B, and Comparative Sample C is supplied with either (i) a "standard" (or "std") biogas feed that contains 55% CH_4, 44% CO_2, and 1% Air (20% O_2, 80% N_2); or (ii) a "high" biogas feed that contains 80% CH_4, 19% CO_2, 1% Air (20% O_2, 80% N_2), as shown in Table 2 (percentages based on volume). The biogas feed (i.e., the raw gas feed) flow is run between normal meter cubed per hour (1,000 Nm^3/hr) to 4,045 1,000 Nm^3/hr, as shown in Tables 2.

For each of Sample A, Comparative Sample B, and Comparative Sample C: (i) the biogas compressed to 11 bar(G) (1100 kPa); and (ii) gas is purged to free air with 0.5% methane loss overall. The results are shown in Table 2.

In Table 2, "RF" refers to the ratio per inlet raw gas. RF is calculated in accordance with the following Equation 1:

$$RF = \frac{\text{Gas Inlet First Compressor (Nm3/hr)}}{\text{Raw Biogas Feed (Nm3/hr)}}. \quad \text{Equation 1}$$

of the same size, relative to an apparatus operating at a higher RF value, which suggests that the apparatus can handle processing a higher total volume of gas. For example, a sample with an RF of 1.4 with a compressor capacity of 1,500 Nm^3/hr can handle 1,071 Nm3/h raw gas (calculated as 1500/1.4=1,071 Nm^3/h), while a sample with an RF of 1.6 can only handle 937.5 Nm^3/h raw gas.

The present disclosure has described one or more preferred embodiments, and it should be appreciated that many equivalents, alternatives, variations, and modifications, aside from those expressly stated, are possible and within the scope of the invention. For example, the structure, arrangement, and elements of the apparatus described herein include alternative design choices and serve the purpose of adapting the apparatus to specific needs, such as specific input compositions or pressures, or specific output purity or yield, for example.

I claim:

1. An apparatus for separating a gas mixture having at least two components, comprising:

a plurality of membrane separation stages including a first membrane stage, a second membrane stage, and a third membrane stage;

TABLE 2

Example 2 Samples Run with FH Membranes													
Sample	S-A1	CS-C1	S-A2	CS-B1	CS-C2	S-A3	CS-C3	S-A4	CS-C4	CS-B2	CS-B3	CS-C5	S-A5
Apparatus Type	App. 10	3TI	App. 10	3T	3TI	App. 10	3TI	App. 10	3TI	3T	3T	3TI	App. 10
Raw Gas Feed (Nm^3/hr)	1000	1000	1000	1000	1000	1035	1040	4000	4000	4000	4000	4020	4045
Biogas Feed Type	std	std	std	std	std	std	std	high	high	high	high	high	high
Compressor 1 Nm^3/hr	1623	1626	1631	1743	1766	1675	1815	6086	5806	5836	6301	6354	6139
Compressor 2 Nm^3/hr	109	N/A	111	N/A	N/A	101	N/A	269	N/A	N/A	N/A	N/A	265
RF	1.62	1.63	1.63	1.74	1.77	1.62	1.75	1.52	1.45	1.46	1.58	1.58	1.52
Nm^3/hr bio/membrane	9.39	9.27	9.27	7.51	7.50	9.61	7.82	14.11	13.30	12.93	10.55	10.99	14.28
Total Product (Nm^3/hr)*	563	563	563	563	562	583	586	3276	3278	3279	3278	3296	3315
Recovery of CH_4* (vol %)	99.50	99.51	99.50	99.51	99.50	99.50	99.51	99.50	99.50	99.50	99.49	99.50	99.50

*In final product, such as from the third retentate stream 32 in Sample A.
N/A = not applicable

As shown in Table 2, Example 2's S-A4 and S-A5 each is run under similar conditions compared to CS-B2, CS-B3, CS-C4, and CS-C5, including the use of high biogas fed at a flow rate of between 4,000 Nm^3/hr and 4,045 Nm^3/hr, yet S-A4 and S-A5 exhibits a higher Nm^3/hr bio/membrane value relative to the comparative samples (CS-B2, CS-B3, CS-C4, and CS-C5).

As shown in Table 2, Example 2's S-A3 is run under similar conditions compared to CS-C1, including the use of standard biogas fed at a flow rate of between 1,000 Nm^3/hr and 1,035 Nm^3/hr, yet S-A2 exhibits a desirable combination of a lower RF value and a higher Nm^3/hr bio/membrane value relative to the comparative sample (CS-C1).

A higher Nm^3/hr bio/membrane value is advantageous because it indicates that fewer individual membranes are needed to obtain the same amount of methane in the final product (i.e., in the third retentate stream). Fewer membranes being included in the apparatus results in reduced economic costs. A lower RF value is advantageous because it indicates that a smaller capacity compressor may be utilized in the apparatus, which reduces the economic cost of the apparatus, reduces the amount of energy required to operate the apparatus, and reduces the physical space of the apparatus. Additionally, a lower RF value indicates that a higher volume of raw biogas can flow through a compressor

the first membrane stage having a first gas separation membrane configured to separate the gas mixture into a first retentate stream and a first permeate stream;

the second membrane stage having a second gas separation membrane configured to separate the first permeate stream into a second retentate stream and a second permeate stream, wherein the second retentate stream is placed in fluid communication with the first retentate stream to form a mixed fluid stream;

the third membrane stage having a third gas separation membrane configured to separate the mixed fluid stream into a third retentate stream and a third permeate stream, wherein the third retentate stream is configured to be withdrawn as a product or discarded; and

a first gas transport device having a suction side connected to the second retentate stream and a discharge side connected to a continuing stream, wherein the continuing stream places the first retentate stream in fluid communication with the first gas transport device.

2. The apparatus of claim **1** further comprising:

a gas source comprising the gas mixture, the gas source configured to be in fluid communication with the first membrane stage.

3. The apparatus of claim **2** further comprising:

a second gas transport device having a suction side and a discharge side, wherein the suction side is connected to an inlet gas line that places the gas source in fluid communication with the second gas transport device, and wherein the discharge side is connected to a feed gas line that places the second gas transport device in fluid communication with the first membrane stage.

4. The apparatus of claim **3**, wherein the third permeate stream of the third membrane stage is recycled and provided to the inlet gas line.

5. The apparatus of claim **3**, wherein the first gas transport device and the second gas transport device comprise a compressor or a blower.

6. The apparatus of claim **3**, wherein the first gas transport device and the second gas transport device are sized such that a mass flowrate of gas transported through the second gas transport device to the first gas transport device is a ratio of 10:1 to 200:1.

7. The apparatus of claim **1**, wherein the first gas separation membrane, the second gas separation membrane, and the third gas separation membrane have a selectivity of carbon dioxide/methane that is greater than 3.

8. The apparatus of claim **3** further comprising:

a feed line measuring device configured to acquire one or more process parameters associated with the inlet gas line and/or in the feed gas line; and

a controller in electrical communication with the second gas transport device and the feed line measuring device, the controller executing a stored program to:

adjust a speed of rotation of the second gas transport device to obtain a desired pressure set-point in the feed gas line, wherein the speed of rotation of the second gas transport device is adjusted in response to a change in the one or more process parameters in the inlet gas line and/or in the feed gas line.

9. The apparatus of claim **8**, wherein the feed line measuring device includes a flowrate measuring device and/or a pressure measuring device and/or a composition measuring device.

10. The apparatus of claim **1** further comprising:

a first retentate measuring device configured to acquire one or more process parameters associated with the first retentate stream;

a controller in electrical communication with the first gas transport device and the first retentate measuring device, the controller executing a stored program to:

adjust a speed of rotation of the first gas transport device to obtain a desired pressure set-point in the continuing stream, wherein the speed of rotation of the first gas transport device is adjusted in response to a change in the one or more process parameters associated with the first retentate stream.

11. The apparatus of claim **10** wherein:

the first retentate measuring device comprises a pressure measuring device, and wherein the controller is further programmed to adjust the speed of rotation of the first gas transport device such that the pressure of the continuing stream is substantially the same as the pressure of the first retentate stream.

12. The apparatus of claim **1** further comprising:

a third retentate measuring device configured to acquire one or more process parameters associated with the third retentate stream; and/or

a second permeate measuring device configured to acquire one or more process parameters associated with the second permeate stream;

a process control device configured to increase or decrease the pressure of the first membrane stage, the second membrane stage, or the third membrane stage;

a controller in electrical communication with the third retentate measuring device, the second permeate measuring device, and the process control device, the controller executing a stored program to:

adjust the pressure of the first membrane stage, the second membrane stage, or the third membrane stage to a desired pressure set-point in response to a change in the one or more process parameters associated with the third retentate stream and/or the second permeate stream.

13. The apparatus of claim **12**, wherein the third retentate measuring device and/or the second permeate measuring device includes a composition measuring device.

14. A process for separating a gas mixture having two or more components, the process comprising:

feeding the gas mixture to a first membrane separation stage, wherein the first membrane separation stage separates the gas mixture into a first retentate stream and a first permeate stream;

separating the first permeate stream into a second retentate stream and a second permeate stream using a second membrane separation stage;

feeding the second retentate stream of the second membrane separation stage into the first retentate stream of the first membrane separation stage to form a mixed fluid stream;

providing a gas transport device having a suction side connected to the second retentate stream and a discharge side connected to a continuing stream, wherein the continuing stream places the first retentate stream in fluid communication with the gas transport device; and

providing a controller in electrical communication with the gas transport device and a first retentate measuring device, the controller executing a stored program to:

adjust a speed of rotation of the gas transport device to obtain a desired pressure set-point in the continuing stream, wherein the speed of rotation of the gas transport device is adjusted in response to a change in one or more process parameters associated with the first retentate stream.

15. An apparatus for separating a gas mixture having at least two components, comprising:

a plurality of membrane separation stages including a first membrane stage, a second membrane stage, and a third membrane stage; and

the first membrane stage having a first gas separation membrane configured to separate the gas mixture into a first retentate stream and a first permeate stream,

the second membrane stage having a second gas separation membrane configured to separate the first permeate stream into a second retentate stream and a second permeate stream, and

the third membrane stage having a third gas separation membrane configured to separate the first retentate stream into a third retentate stream and a third permeate stream, wherein the second retentate stream is placed in fluid communication with the third retentate stream to form a mixed retentate stream;

wherein the second retentate stream is split into a first continuing stream and a second continuing stream, wherein the first continuing stream is placed in fluid communication with the first retentate stream, and the second continuing stream is placed in fluid communication with the third retentate stream to form the mixed retentate stream.

16. The apparatus of claim 15, wherein a valve is configured to regulate the flowrate between the first continuing stream and the second continuing stream.

17. The apparatus of claim 16 further comprising:

a controller in electrical communication with the valve, the controller executing a stored program to:

adjust the flowrate or pressure of the first continuing stream and the second continuing stream by moving the valve between an open and closed position.

18. The apparatus of claim 17 further comprising:

a third retentate measuring device configured to acquire one or more process parameters associated with the third retentate stream,

wherein the controller is in electrical communication with the third retentate measuring device, the controller executing a stored program to:

adjust the flowrate or pressure of the first continuing stream and the second continuing stream based on the one or more process parameters in the third retentate stream.

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